Strongly Correlated Electrons and Neutron Scattering

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Various aspects of close and interesting relationship between antiferromagnetism and singlet ground states are introduced for which neutron scattering have been playing vital roles. The special emphasis is on the disorder-induced antiferromagnetism in spin-Peierls systems, which can be viewed as a nucleation process of classical magnetic order in the background of singlet state whose origin is purely quantum-mechanical. It is then pointed out that similar features will be found in $Ce_xCu_2Si_2$ and high T_c cuprates. Finally the possible charge ordering process in NaV_2O_5 is discussed which leads to the quenching of localized spins.

KEYWORDS: A. Magnetic Materials, Superconductors, D. Magnetic Properties, Phase Transitions, Superconductivity

§1. Introduction

Interactions between electrons in solids result in various states of matter and phase transitions. Coulomb repulsive forces are known to be the cause of magnetism. Especially in the presence of strong Coulomb interaction, i.e. in strongly correlated systems, Mott insulators are realized in the half-filled non-degenerate band, or in the case with integer number of electrons per site in general, where the low lying excitations are exclusively due to spin degrees of freedom. This situation is properly described by the Heisenberg spin Hamiltonian. Here spins and charges of electrons are completely separated. Although various types of magnetic ground states have been explored in a transparent way by this Heisenberg spin Hamiltonian, much of the recent interest are in the nonmagnetic singlet ground states, which are usually termed as spin-gapped systems. These singlet ground states are purely quantum-mechanical in their origin in comparison to magnetic states which are basically classical. Such studies on singlet ground states may be motivated by the fact that the high temperature superconductivity in cuprates, which has singlet $d_{x^2-y^2}$ symmetry, are realized next to AF Mott insulators, i.e. in the doped Mott insulators where small amount of holes are introduced into the Mott insulating state[1)]. Here it is to be noted that AF superexchange interaction, J, has two distinct aspects; it leads quite generally to AF Néel ground states in lattices,

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while it results in the singlet states obviously for two spins and even in lattices in the presence of strong quantum fluctuations or frustrations. Actually it turned out that there exists a close relationship between antiferromagnetism and singlet $d_{x^2-y^2}$ superconductivity[2]. In this paper some of the recent development of the studies on the remarkable features of this interrelationship between antiferromagnetism and singlet ground states are introduced where neutron scattering experiments have played decisive roles.

§2. Disordered Spin-Peierls Systems

The spin-Peierls transition has been known for some time[3,4] but the discovery of this transition in inorganic compound, $CuGeO_3$, by Hase *et al*[5] is rather recent. Surprizing and puzzling was the report of the neutron scattering experiment by Regnault *et al*[6] on $CuGeO_3$, with a small amount of replacement of Ge by Si. This experiment has revealed the coexistence of resolution-limited sharp Bragg spots, one corresponding to the lattice dimerization stabilized below around the critical temperature of spin-Peierls transition of the parent clean systems and the other to AF ordering stabilized at much lower temperatures. Since these two states, dimerized and AF states, had been considered to be exclusive[4], some kind of inhomogeneity was suspected. At the same time, the sharpness of these two Bragg spots together with the apparent transfer of the spectral intensity between these two spots as the temperature is varied had suggested that the experiment had in fact disclosed the coexistence of the true long range order of these two conflicting order parameters. A theoretical proposal[7] has been made, which indicates that such coexistence is possible once the spacial variations of the competing order parameters are taken into account. In this study the lattice distortions are treated completely classical because of the three-dimensionality of the actual crystals and the quantum mechanical features of one-dimensional spins are expressed in terms of the phase Hamiltonian[8] derived by the bosonization[9], which has been treated in a mean field approximation in view of the existence of the interchain exchange interaction. Basic physics behind turns out to be that the realization of the singlet ground state by the dimerization is purely quantum mechanical and the complete coherency of the wave functions of spins in whole crystal (of the order of 10^{23} !!) is needed and that, once the quantum phases of wave functions are perturbed, magnetizations are created locally, which naturally order in the absence of frustrations. The excitations above this new type of ground state with the coexistence turned out to have unique features[10]: i.e. there are two distinct excitations, one with a gap reflecting dimerization which is present even in the clean systems and the other at very low energy and at around AF wavevector only with the very little total spectral weight in proportion to the degree of disorder,i.e. impurity concentration. In the coexisting ground state this small spectral weight at low energy forms well-defined spin wave mode reflecting the long range AF order. This is schematically shown in Fig.1.

Such a feature of two-mode structure has actually been observed in neutron scattering experiment [11,12] and the existence of the well-defined spin waves has also been proven by the ESR experiments

as well[13,14]. At present the onset of the Néel ordering has been observed even down to 28.5mK at $x = 5 \times 10^{-3}$ of $Cu_{1-x}Zn_xGeO_3$ by Manabe *et al.*[15]. Note that there is little weight, i.e. "transparent", in the energy region between the gap and spin wave mode. This is very unusual and unexpected since the effects of perturbations had been expected to be treated "perturbatively" in the presence of the gap in the excitation spectrum and then only the modifications of the gapped modes could be expected. At the same time this finding that the disorder introduces spectral weight only at very low energy (as low as the elastic Bragg spots) even in the presence of large gap will be a warning to correlate the experimental data of neutron scattering and NMR without serious considerations on the sample quality, since only the NMR will be affected by such low energy excitations. The present problem of disorder-induced antiferromagnetism in spin-Peierls systems can be considered to be a typical example showing how the spectral weight of the excitations emerges in the process of nucleation. Here the first order transition is expected in the clean systems between AF ordering and spin-Peierls dimerized state as a result of the competition between the interchain exchange interaction and the spin-lattice coupling along the chain[4,16]. Once the disorder is induced, however, the antiferromagnetism is nucleated around the impurities in otherwise singlet ground state.

Fig. 1. A schematic representation of the distribution of the spectral weight in the disordered spin-Peierls systems. Note that disorder introduces small but finite amount of spectral weight at very low energy only at AF wavevector.[10]

§3. Heavy Electrons, $Ce_xCu_2Si_2$

Among strongly correlated electron sysytems, heavy electrons in lantanides and actinides have been studied for a long time and it is now known that magnetism and superconductivity are next to each other. Very recently Ishida *et al*[17] found very mysterious properties in Cu-NQR spectra of

series of $Ce_xCu_2Si_2$. At x=0.975 this system is AF ordered, while at x=1.025 the heavy electron superconductivity is observed. However at intermediate x=0.99 they found that there exist magnetic fluctuations with very low frequencies comparable to the NQR frequency (ω=$10^{6\sim7}$)Hz even though no signatures of magnetism are seen in neutron scattering whose characteristic frequency is ω=$10^{11\sim12}$Hz. Since there should be intrinsic disorder due to the spacial distribution of Ce atoms, a similar consideration as in the preceding disordered spin-Peierls systems will apply and then a similar feature as in Fig.1 is expected though the gapped mode here is due to superconductivity instead of dimerization in disordered spin-Peierls systems.

§4. High T_c Cuprates

Disclosures of the true nature of magnetic exciatations have played crucial roles in the understanding of high T_c cuprates, for which both neutron scaterring and NMR have contributed very much. Findings by the former will be fully discussed in this Symposium. On the other hand NMR experiment on the undredoped Y(123) by Yasuoka *et al.*[18] has first indicated the existence of the spin gap: one of the most remarkable features of the cuprates. All these experimental studies on magnetic excitations have revealed that the disorder affects the excitations in essential ways as in the case of disordered spin-Peierls systems. Few of the examples will be discussed in the following. First the complete destructions of the spin-gap in NMR by a very small amount of replacement of Cu by Zn in Y(248)[19] will be understood again by Fig.1 if the spin-gap is associated with the formation of the short range order of spin-singlet as described by the RVB theory based on the slave boson mean field approximation of the *t*-*J* model[20] as shown in Fig.2[21]. Here only the general trends of the spin-charge separation and their confinements[22] are stressed. This spin gap can be identified with the pseudogap observed later by ARPES experiments[23, 24] since electrons are convolutions of spinons and holons[25], as has been explicitly demonstrated[26]. In this context the underdoped region of $La_{2-x}Sr_xCuO_4$ is very interesting. Early neutron scattering data by Keimer *et al*[27] indicated the existence of spin glass phase, which at the same time has a feature close to the long-range ordered antiferromagnetgism of parent compounds. This finding has led the present author to conjecture that the true long range AF ordering might actually been realized at lower temperatures[28]. However more recent neutron studies[29] unambiguously demonstrated that the ground state is a canonical spin glass phase. A possible cause for this stability of spin-glass over the long range AF ordered state may be due to the extended nature of the wave function of the holes, which will lead to the frustration among spins[24]. This difference between extended and local disorder is obviously dependent on the mobility of holes, i.e. insulating or (weakly) conducting, and may be related with the experimental findings of shift of the spectral weight toward the low energy in accordance with the resistivity increase as the temperature is lowered in underdoped YBCO[30, 31].

Fig. 2. A schematic representation of the phase diagram based on the slave boson mean field theory of the t-J model. The spingap, which is the same as the pseudogap in this theory, is associated with onset of singlet RVB order parameter.[26)]

§5. $\mathbf{NaV_2O_5}$

This system, which is insulating at all temperature, exhibits a magnetic transition at $T_{\rm c} = 35K$ below which the spin susceptibility drops sharply but continuously[32)], a feature typical of spin-gap systems, and has been believed to be an example of spin-Peierls transition.In this case the valences of V atoms are considered to be arranged as in Fig.3(b). However recent NMR experiment on V atoms by Ohama *et al*[33)] indicated that the charge disproportionation sets in below $T_{\rm c}$ even though the the charge distribution on V atoms is uniform above $T_{\rm c}$. This finding stimulated many studies both theoretical and experimental. A theoretical study[34)] based on the selfconsistent

Fig. 3. A possible spacial pattern of charge ordering in NaV_2O_5.[34]

Hartree approximation for both onsite and intersite Coulomb interaction has disclosed that the ground state will have a charge ordering of zigzag type as seen in Fig.3(c), which has a spin dimer structure. Above $T_{\rm c}$ the charge distributions are expected to be as in Fig.3(a). Basically all experimental data appear to be consistent so far with this state, including the spin excitation spectra probed by the neutron scattering by Yosihama *et al*[35,36]. It will be very interesting to probe the transient region of this charge ordering which leads to the formation of the singlet ground state, i.e. the quenching of the quantum spins.

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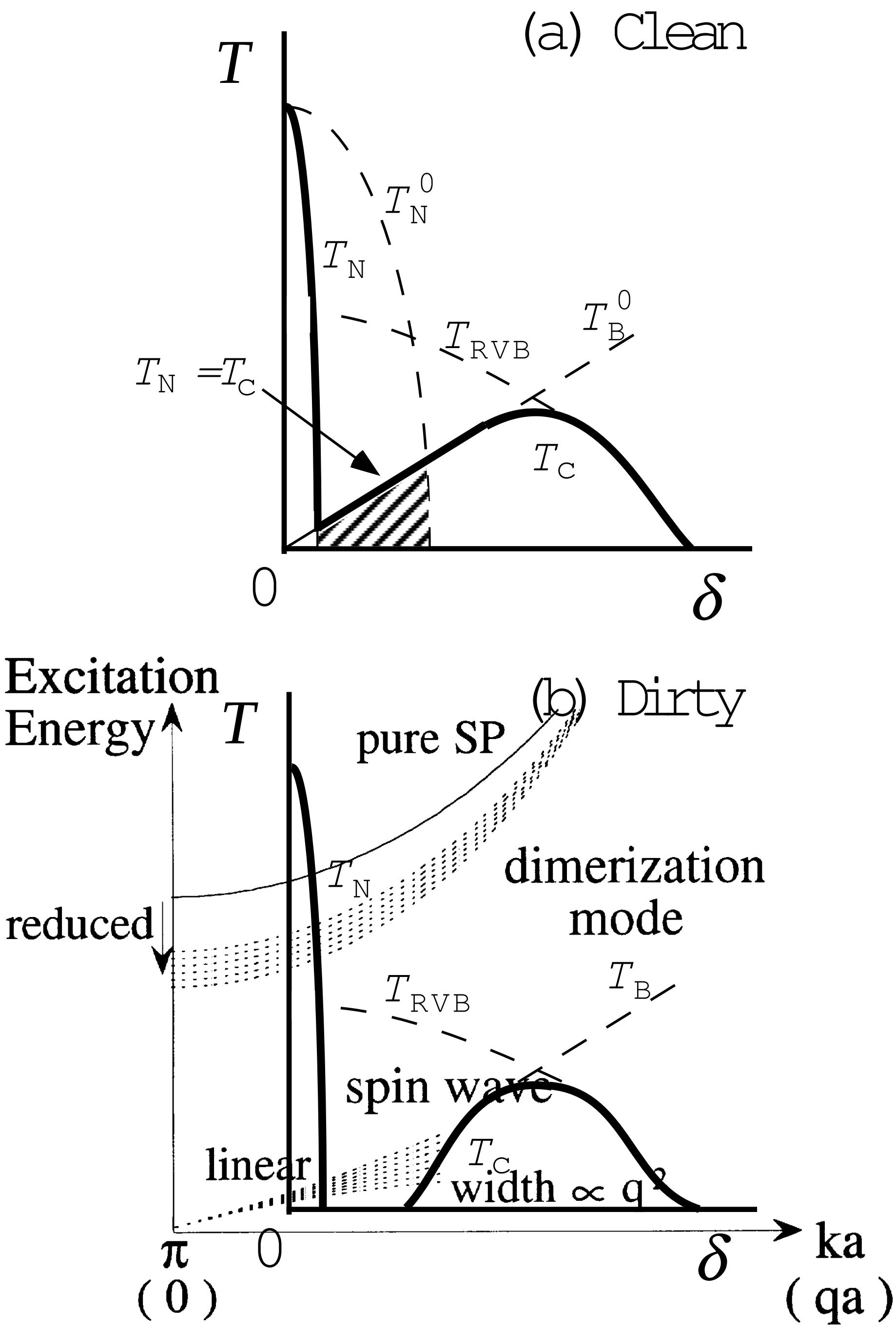
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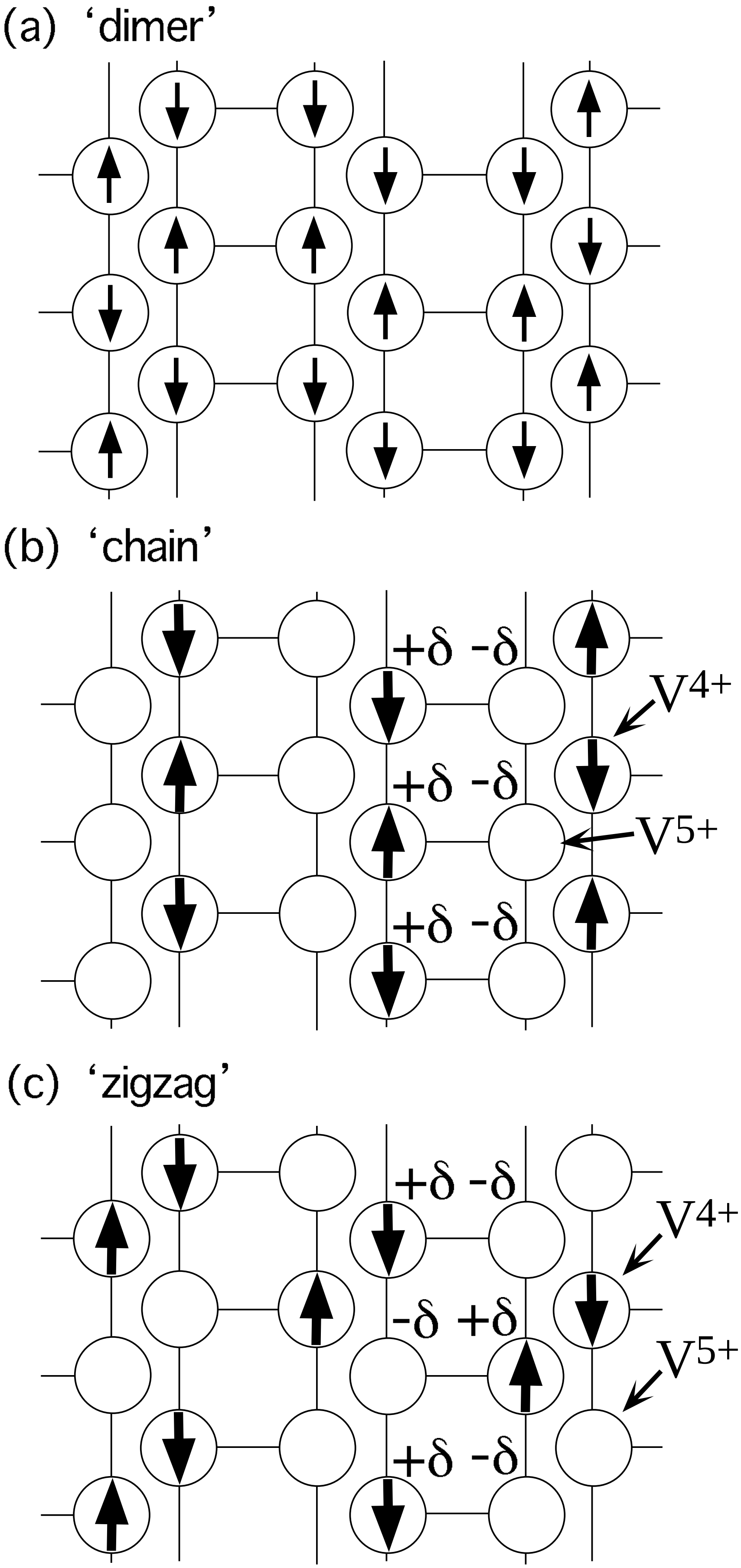
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